Mid-Infrared Third-Harmonic Emission from Heavily-Doped Germanium Plasmonic Nanoantennas

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Abstract: We investigate the nonlinear optical properties of single resonant plasmonic antennas fabricated from heavily-doped Germanium films. Excitation with intense and ultrashort mid-infrared pulses at 10.8 μm wavelength produces emission at 3.7 μm via third-harmonic generation.

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Recent advances in semiconductor film deposition allow the growth of heavily doped germanium with effective plasma frequencies of up to 60 THz, corresponding to 5 μm wavelength. This technology paves the way for mid-infrared (MIR) plasmonics with application to integrated telecommunications systems and to precise sensing in the spectral region defined as the vibrational fingerprint of molecules. Characteristics like CMOS compatibility, low electron effective mass and tunable dielectric function give significant advantage to Ge over other semiconductors [1,2]. In addition, metals like gold display relatively poor plasmonic performances in the MIR due to a disadvantageous dielectric response that leads to weak confinement of the electromagnetic field.

In this work, we demonstrate that plasmonic Ge antenna structures are also suitable for driving nonlinear optical processes such as third-harmonic generation (THG) in the mid infrared [3] owing to the strong near-field enhancement of the light-matter coupling. These devices act as light emitters constrained at sub-wavelength dimensions and consequently are of high interest for experiments targeting single molecules or other isolated quantum systems at ultrashort timescales [4].

Doped single-crystalline germanium films with a thickness of 1 μm are grown epitaxially via low-energy plasma enhanced chemical vapor deposition (LEPECVD) on intrinsic silicon substrates. Annealing steps ensure the highest dopant activation. Resonant antenna structures including double rod antennas (see Fig. 1(a)) are then fabricated via electron beam lithography. Simulations considering the antenna geometry and the material dielectric function close to the plasma edge determine the exact resonance for mid-infrared excitation [5]. The optical characterization system is

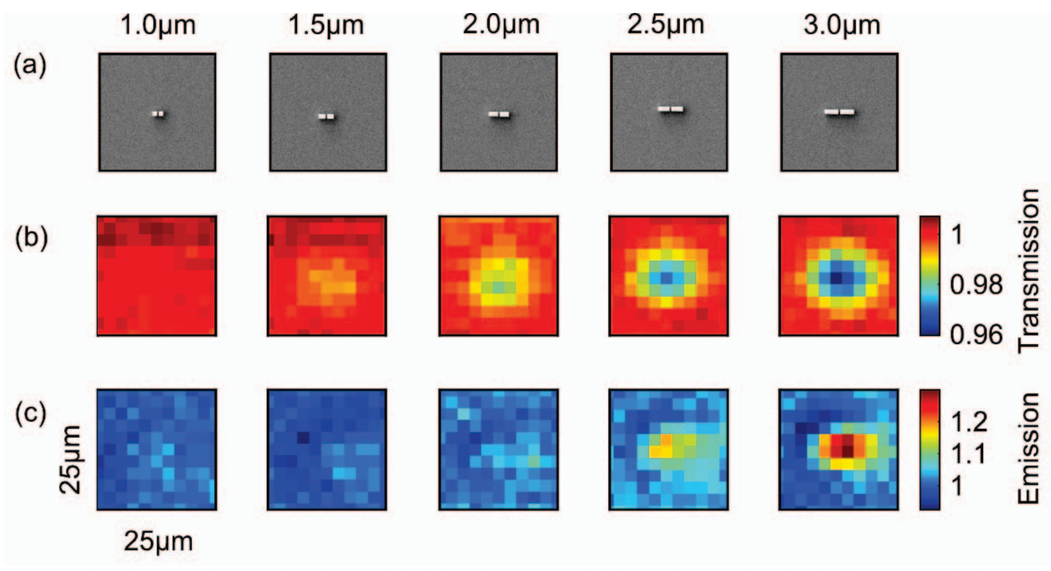

Fig. 1. (a) Scanning electron micrographs of several double rod antennas with increasing arm length; (b) linear transmission maps of each double rod antennas illuminated with a laser pulse centered at the wavelength of 12 μm; (c) spatially resolved third harmonic emission normalized to the silicon substrate background. The dimension of each panel is 25μm by 25μm.

pumped by a Yb:KGW femtosecond laser that drives the nonlinear frequency conversion stages for the generation of intense few-cycle pulses tunable in the mid-infrared spectral range from 8 μm to 22 μm wavelength. Excitation fields of up to 20 MV/cm are reached in the focus of home-made microscope equipped with a dispersion-free Cassegrain-Schwarzschild reflecting objective (NA=0.5). A second, identical objective is used to image the nanostructure in transmission geometry. Dielectric and semiconductor filters as well as a grating monochromator are employed for spectral filtering.

A liquid nitrogen cooled mercury cadmium telluride detector collects the emission while the sample is scanned through the common focus. This setup allows the addressing of single antennas and the linear and nonlinear cross-sections to be mapped. We study different antenna geometries, sizes and material doping levels.

Fig. 1(b) demonstrates the transmission images at the fundamental wavelength with the increased scattering of isolated nanostructures while Fig. 1(c) plots the corresponding THG emission that is strongly enhanced at the antenna position with respect to the substrate. Fig. 2(a-b) demonstrates the spectral characterization of the third harmonic photons that are generated at 3.7 μm in comparison to the fundamental excitation set at 10.8 μm.

The experimental variation of the excitation power reveals the nonlinear dependence (Fig. 2(c)) typical for third-order optical processes.

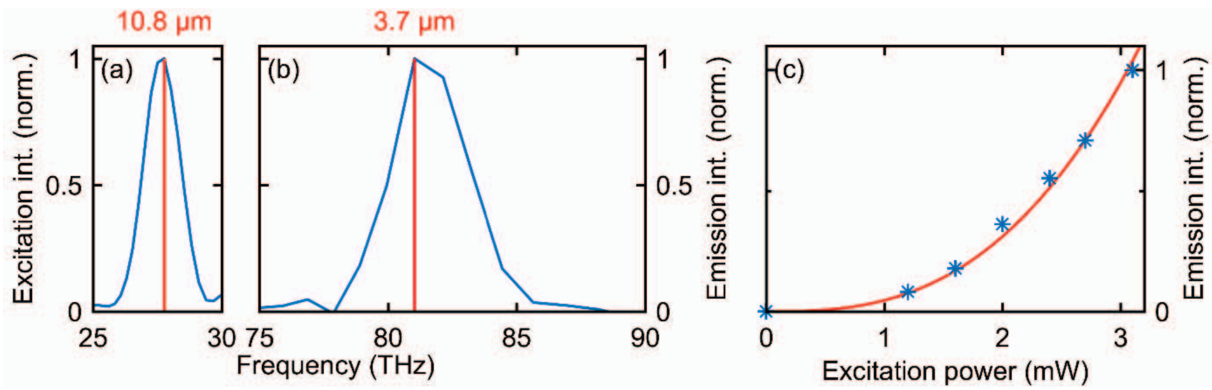

Fig. 2. (a) Normalized excitation and (b) THG emission intensity spectra from a germanium nanoantenna; (c) power dependence of the nonlinear emission of a single Ge antenna (blue: measurement, red: fitting curve ~ $x^{2.8}$).

In conclusion, we demonstrate third-harmonic generation from heavily doped Ge structures illuminated by intense excitation with femtosecond pulses owing to the plasmonic enhancement occurring in the near-field. Such nonlinear optical antennas are sources of ultrashort radiation constrained below the diffraction limit. This work opens interesting perspectives for experiments targeting molecular resonances or other isolated quantum systems in the MIR frequency range.

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